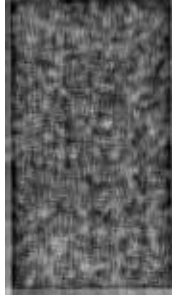


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A Literature Survey

Research and
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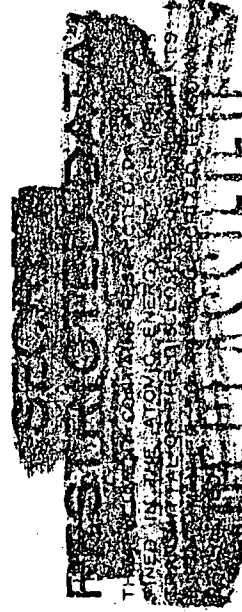


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THE RELEASE OF FISSION PRODUCTS FROM REACTOR FUEL

A Literature Survey

P. H. WILKS

Radiation and Criticality Safety

March 23, 1959

United States Air Force

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ABSTRACT

A discussion of available data on fission product release is presented. The survey is divided into data from actual incidents, field release experiments, laboratory release from fuel element samples, and pyrometallurgy. An author index and an alpha-numerical index are included.

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INTRODUCTION

The accidental release of fission products from an operating nuclear reactor represents a potential hazard to those nearby. For this reason, a summary of helpful information on tests, ideas and experience related to fission product release would be of value to persons involved with reactor safety. This survey has been prepared to fill that need, but relevant data are limited. The need for more research in this field is apparent.

It is to the credit of the industry that incidents involving fission product release are few, and much of the available information dealing with large scale release is theoretical in nature. A very limited number of field release experiments has been performed. Laboratory scale experiments have provided some data regarding the fractions of various fission products which are released when fuel or fuel elements are melted, but the applicability of these data toward predicting the seriousness of full scale reactor incidents is open to question.

The following is a survey of the literature which is related to the general problem of fission product release from operating reactors. It is separated into four categories according to the type of information offered by each reference. The categories are as follows: (I) Fission Product Release from Actual Incidents; (II) Field Release Experiments; (III) Laboratory Scale Release from Fuel Element Samples; and (IV) Pyrometallurgy of Spent Uranium Fuel. An alphabetical author index and an alpha-numerical index are also included.

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This survey was prepared using the CE-ANPD library card files, Nuclear Science Abstracts, and Abstracts of Classified Reports as primary sources of references. The subject headings which were explored are as follows:

FISSION PRODUCTS

Escape

Release

URANIUM

Fission Products

Purification

HIGH TEMPERATURE SEPARATION

Processes

Pyrometallurgy

Fuel Elements

Separation Processes

REACTORS

Safety

Disasters

Nuclear Science Abstracts was explored from Volume I to Volume XIII, No. 1. Abstracts of Classified Reports was consulted from Volume I to Volume XV, No. 1. In addition, Chemical Abstracts was investigated from 1950 to 1958 inclusive.

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The names of research workers attending a meeting of Fission Product Release Studies at Germantown, Maryland, November 6, 1958 and some of their subordinates were sought in the "Authors" card catalogue. The quarterly reports of their respective facilities were sought in the "Corporate Entries" card catalogue.

In December of 1953 a working party at Harwell, England published plans for a series of fission product release studies. The names of all members of this group were checked in the "Authors" catalogue.

Additional information was obtained through personal communications with C. C. Gamertsfelder and R. E. Baker of Radiation and Criticality Safety, GE-ANP; H. M. Feder and N. R. Chellew of Argonne National Laboratory; G. E. Creek of Oak Ridge National Laboratory; H. G. Hembree, G. V. Beard, and J. Davenport of the AEC, Washington, D. C.; R. C. Brothers of the AEC, Pembroke, Ontario; and J. E. Woolston of Atomic Energy of Canada Limited.

An attempt has been made to restrict the references to those offering data which might conceivably be of value in predicting fission product release caused by overheating in an actual reactor. While the amount of information in this survey is not great, it is felt that the picture of the available data on fission product release which it presents is as complete as is possible at the time of this writing.

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(I) FISSION PRODUCT RELEASE FROM ACTUAL INCIDENTS

In December of 1952, an excursion occurred at Chalk River, Ontario which resulted in 10,000 curies of long lived fission products being leached by cooling water from damaged fuel elements.^(1,2) The contaminated water was confined and very little airborne radioactivity was released from the facility. Approximately eleven percent of the fuel elements were sufficiently overheated to melt and thereby rupture the sheathing. A less serious accident occurred at the NRTS in February, 1957 involving release of fission products to cooling water of the MTR.⁽³⁾

The accident at the Windscale No. 1 pile in October of 1957^(4,5) resulted in appreciable I^{131} contamination of the countryside and distribution of some locally produced milk was stopped. The reactor released 5×10^4 curies of I^{131} , of which 3×10^4 curies passed through the filtration system and into the atmosphere. Estimates place the total deposition of I^{131} on surrounding vegetation at 2×10^4 curies. The accident caused 3-1/2 tons of uranium to be oxidized while the total iodine release to the filtration system represents that from 2-1/2 tons of uranium.⁽⁶⁾ It may be inferred from this that the fractional release of iodine from the affected area was 0.7.

A power excursion in an ANP reactor in November, 1958 resulted in the release of about one tenth percent of the total gross fission products in the reactor.⁽⁷⁾ Actual measurements of the amounts of fission products released are presented in Table I.⁽⁸⁾ The fuel elements in this reactor were of the metallic

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type, having a core of UO_2 in intimate mixture with powdered 80 Ni-20 Cr and clad with 80 Ni-20 Cr.

Table I

Fission Product Release from ANP Excursion, November, 1958

Reactor Inventory-Curies	<u>STACK RELEASE</u>			
	From Stack Monitors		Inferred from Deposition on Vegetation	
	Curies	F	Curies	F
Gross (at 10 min) 4×10^5	400	0.001	---	---
I^{131} (at 0 min) 80	0.16	0.002	0.3	0.004
I^{133} (at 0 min) 480	3.2	0.0067	4	0.0083
I^{135} (at 0 min) 1100	27	0.025	30	0.027
Sr^{91} (at 0 min) 900	0.58	0.0006	1	0.001

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(II) FIELD RELEASE EXPERIMENTS

In May, 1958, an experiment was performed at GE-ANPD's Idaho Test Site to determine the effect of burnout of one metallic fuel tube of a high performance air cycle reactor.⁽⁹⁾ Burnout was accomplished by reducing the flow of cooling air to the tube. The fractional release of I^{131} ranged from 0 to 0.7 during this experiment, depending on the thermal damage to each portion of the cartridge. Fractional release for the total cartridge was about 0.25.

The Nuclear Aircraft Research Facility, operated by Convair - Fort Worth, has completed a series of fission product release experiments using GE-ANP metallic fuel elements. The fuel samples were heated by induction in graphite crucibles and the distribution of the resulting fallout was studied along with the fractional release of fission products. While analysis of the data is not complete at the time of this writing, descriptions of initial plans and a preliminary discussion of the results are available.^(10,11,12)

Chamberlain and McGaw of Harwell have performed field release tests in England using I^{131} , but no details of this work have as yet been made available to the U. S. AEC.⁽¹³⁾

In 1953, a reactor designated as BORAX-1 was intentionally destroyed by completely ejecting a control rod worth 4% K-eff and allowing the fuel to melt.⁽¹⁴⁾ The ensuing reaction between molten fuel and reactor water threw most of the contents of the shield tank into the air. Practically all of the fuel

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originally in the reactor was found within a radius of 350 feet around the reactor. Gamma dose rates to the surrounding terrain were quite small even at a distance of less than a mile downwind from the reactor. A detailed report dealing with the health physics aspects of this experiment is available.(15)

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(III) LABORATORY SCALE RELEASE FROM FUEL ELEMENT SAMPLES

Parker and Creek of ORNL have contributed most of the available experimental data on fission product release from controlled melting of irradiated fuel specimens. Their data are reproducible but are from very small scale experiments.^(16,17) At trace concentrations of fission products, slow melting of APFR type stainless steel plate at 1525°C in air or steam released half of the rare gases, one third of the iodine, one tenth of the cesium and traces of strontium. After 25% burnup the fraction of cesium released increased to 0.6. MTR type aluminum clad samples, containing traces of fission products, released 0.02 of the iodine and 0.10 of the rare gases upon melting at 700°C. Zircaloy type fuel specimens, melted at 1850°C in helium after 15% burnup, released up to 0.95 of the rare gases, 0.6 of the cesium, and 0.80 of the iodine. In air at comparable temperatures the iodine release was reduced to 0.53 and the cesium to 0.05. Strontium release was less than 0.01 in all cases.

Experiments where Parker and Creek employed postage stamp size GE-ANPD metallic fuel specimens have been reported in their latest work.⁽¹⁷⁾ Melted rapidly and cooled immediately, they released 0.10 of the xenon, 0.04 of the iodine, traces of cesium and strontium. Another specimen, kept at a temperature above the melting point for four hours, oxidized completely and released 0.77 of the xenon and krypton, 0.99 of the iodine, 0.016 of the cesium and a trace of strontium. The investigators themselves question the validity of the iodine value of 0.99, there being evidence that it is actually closer to 0.77.⁽¹⁸⁾ Parker, Creek, and Martin have recently performed some experiments with UO₂ type fuel. Their preliminary data indicate oxides to be inherently retentive, since it was found

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that release of volatile fission products at 1600°C is considerably less than would be expected from metallic fuel. In two experiments, rare gas release fractions of 0.01 and 0.013 were obtained.

Lustman and others^(19,20) have studied the evolution of rare gases from irradiated UO_2 in terms of diffusion coefficients. Although there is a good deal of scatter in the data, the importance of both porosity and temperature is clear from these studies.

Some experiments have been performed on fission product release from GE-ANPD tubular metallic and ceramic fuel element specimens. The data from these tests, performed in the LITR reactor at Oak Ridge and the MTR in Idaho, are very difficult to analyze. However, some results are available.^(21,22,23,24,25,26) The data are presented in terms of release rate, defined as the percent of nuclei formed which escape to the cooling air stream. With few exceptions release rates were less than 0.1 percent. For metallic samples the most important factors controlling release rate of iodine seem to be integrity of the cladding and temperature. Comparison of data from ceramic elements showed that an increase in temperature of 300°F increased iodine release by a factor of twenty. As might be expected, the porosity of ceramic specimens is another very important factor. Some data have been obtained from coated ceramic samples, but the results are inconclusive at present.

Data were obtained at MSA Research⁽²⁷⁾ by melting Zircaloy clad FWR type elements at 1650°C in a steam atmosphere. The assemblies, which contained only

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trace amounts of fission products, lost about half of the xenon and from 0.01 to 0.26 of the iodine. Iodine release was greater during slow meltdown, where 8 minutes were required to melt the element vs. 4 minutes for fast meltdown.

Hilliard of GE-Hanford Atomic Products Operation has recently published a paper on oxidation of Uranium.⁽²⁸⁾ This work was performed in preparation for future research on fission product release.

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(IV) PYROMETALLURGY OF SPENT URANIUM FUEL

Much work has been done on the purification of irradiated fuel by melting the fuel under various conditions. Since under these conditions volatile fission products escape, this work is somewhat applicable to the problem at hand.

Historical background for pyrometallurgical purification is presented in a literature survey by E. S. Bomar.⁽²⁹⁾ A bibliography on the effect of heating irradiated uranium has been compiled by Hilliard.⁽³⁰⁾

In the laboratory, Whitman⁽³¹⁾ has performed experiments in zone melting in an attempt to remove fission products from uranium, and Gilman⁽³²⁾ removed essentially all the xenon by transient heating of the fuel. Saul⁽³³⁾ heated small (2-4g) surface oxidized fragments of irradiated uranium which had been cooled 3 years to 1275-1400°C. At 1275°C 1/2 of the rare earths and 1/3 of the strontium and cesium migrated to the oxide. At 1400°C 2/3 of the rare earths, 1/2 of the cesium, 1/3 of the strontium and small amounts of plutonium were removed.

Feder and Chellew, however, have gathered the data which are of greatest general interest.^(34,35) Their procedure involves induction heating of "Fissium" (an alloy of uranium with small amounts of molybdenum, ruthenium, zirconium, etc., which is intended to represent spent uranium fuel). Non-radioactive xenon and iodine are added in the proper amounts when it is wished to study the release of these and the addition of irradiated uranium pins furnishes tracer quantities

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of radioactive fission products to indicate the distribution of the non-radioactive atoms.

The following general conclusions can be drawn from the pyrometallurgical data, although their significance with regard to reactor incidents is subject to question.

A very important factor in the fractional release of rare gases is their initial concentration. When concentrations are on the order of 0.01 ppm, escape of xenon takes place by a slow diffusion process. Diffusion is the mechanism of release even at 1300°C, 200°C above the melting point of fissium. However, when concentrations approach those that would be expected in spent fuel (200 ppm), essentially all the xenon is released just above the melting point of 1100°C.

Feder and Chellew have shown that the fraction of iodine released is more dependent on temperature than on concentration. In experiments at 1300°C, one third to one half of the iodine was released over a three hour period in each case even where concentrations were as far apart as 0.008 ppm and 415 ppm. At 1400°C, however, 0.85 of the iodine was released in 2-3 hours from samples containing 3.5 and 200 ppm of iodine. The iodine that had been added was shown to have reacted with metallic uranium to form UI_3 .⁽³⁶⁾ The stability and non-volatility of this compound is thought to have kept to a minimum the dependence of fractional release on initial concentration.

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